REMARKS

This Amendment is made in response to the Advisory Action mailed on May 11, 2001 and is in further response to the Final Office Action mailed on October 12, 2000, and is supplemental to the Amendment After Final Action dated April 12, 2001 (which is incorporated herein by reference). Reconsideration and withdrawal of the objections to and rejections of this application are respectfully requested in view of the amendment and remarks which place the application into condition for allowance or in better condition for appeal.

In light of the commercial importance of this invention, the Examiner was contacted by the undersigned on June 21, 2001. Exercising her discretion, the Examiner refused to discuss this application. The Examiner reasoned that that the quarter had already ended, that Applicants missed their opportunity to interview the Examiner earlier, and that prosecution was, thus, closed. It is clear from the Advisory Action, however, that the Examiner may have misread the cited references and failed to fully appreciate the scope of the instant invention. This Supplemental Amendment is submitted, therefore, to address the Examiner's concerns and clarify any remaining issues she may have.

Claims 16-19 and 21-25 are pending and at issue. Claim 20 is canceled, and claims 16-19 and 22-24 amended, without prejudice, admission, surrender or with any intention to create any estoppel as to equivalents. Applicants reserve the right to pursue canceled subject matter in a continuation application.

No new matter is added.

It is submitted that these claims are patentably distinct from the references cited in the Office Action, and that these claims are in full compliance with the requirements of 35 U.S.C. §112. The amendments to the claims and remarks made herein are not made for the purpose of

patentability within the meaning of 35 U.S.C. §§ 101, 102, 103 or 112; but rather the amendments and remarks are made simply for clarification and to round out the scope of protection to which Applicants are entitled.

The Advisory Action initially notes that the claims include seven multiple dependent claims. (Advisory Action, at 2). The cancellation of claim 20, without prejudice, has reduced the number of multiple dependent claims, thereby also reducing the total number of claims under consideration. The cancellation of claim 20 has also rendered moot the concern in the Office Action that "monodisperse state" is allegedly not disclosed in the specification.

It is also alleged in the Advisory Action that new matter has been introduced. The claims have been amended to remove the recitation "about," thereby mooting the allegation.

Claims 17, 18 and 22 stand rejected under 35 U.S.C. §112, first and second paragraphs, with respect to the recited intrinsic viscosity and heat distortion temperature along the lines set forth in the Final Office Action.

Applicants initially note that the recitation in claim 22 to polyolefin refers to the polyolefin recited in any one of claims 16, 17 or 18.

With respect to the alleged lack of disclosure concerning the conditions under which the intrinsic viscosities and heat distortion temperature are determined, Applicants reiterate their position that the issue is not that standards can change over time (as asserted in the Advisory Action) but what did a skilled artisan know at the time of filing. The law is very clear that an Examiner must inquire as to the knowledge of the skilled artisan at the time of filing of the application. *See In re Epstein*, 32 F.2d 1559, 1564 (Fed. Cir. 1994) ("The time relevant to the level of skill inquiry is when the application was filed[.]"); *see also, Graham v. John Deere*, 383 U.S. 1, 17, (1996) (finding that skill level is measured at the time the invention was made).

Applicants respectfully assert again that a skilled artisan, at the time of filing of the instant application, would readily understand that measurement of the viscosity are routinely determined by the standards set forth in the specification and would, in turn, know which standard to use and what experimental conditions to apply. A skilled artisan would, consequently, not be required to perform any undue experimentation in order to practice the instantly claimed invention in general, or to calculate the intrinsic viscosity of the polyolefin in particular. However, in order to substantiate Applicants' position beyond any shadow of a doubt, Applicants respectfully submit copies of the following Standard Test Methods:

(1) Deflection Temperatures of Plastics Under Flexural Load (ASTM D 648-96) (Exhibit 1);

(2) Dilute Solution Viscosity of Ethylene Polymers (ASTM D 1601-86 Reapproved 1991) (Exhibit 2);

(3) Determination of temperature of deflection under load (ISO 75-2: 1993(E)) (Exhibit 3); and

(4) Determination of viscosity number and limiting viscosity number (ISO 1628-3: 1991/cor. 1: 1992(E)) (Exhibit 4).

Consequently, the objection is obviated.

The Advisory Action further asserts that the specification fails to define the version of the DIN53461 standard submitted in response to the Final Office Action. As noted above, however, a skilled artisan would, by definition, know how to determine the HDT, or for that matter, intrinsic viscosity, and know the experimental protocol and conditions necessary to achieve it. Consequently, any hypothetical change of, or revisions to, DIN 53461 would be irrelevant because, at the time of the filing of the instant application, a skilled artisan would

readily understand that DIN 53461 (which is also readily available to a skilled artisan) could be used to practice the instantly claimed invention. Further, Applicants will submit a declaration substantiating that the version submitted to the Examiner is analogous to the version cited in the specification. As discussed in the previous paragraph, Applicants herein provide copies of other standards available to a skilled artisan for the practicing of the instant invention to further substantiate Applicants' position.

Claims 23 and 24 stand rejected under 35 U.S.C. §112, second paragraph. Claims 23 and 24 have been amended, thereby rendering the rejection moot.

In view of the foregoing, reconsideration and withdrawal of the objections to the specification and rejections under 35 U.S.C. §112, first and second paragraphs, and favorable consideration of the pending claims are respectfully requested.

The Advisory Action maintained the rejection of the claims under 35 U.S.C. §102(b) as being anticipated by Japanese Patent No. 58-149060 ("JP '060"). JP '060 is said to relate to a toner that includes a colorant, carbon black, a charge controlling agent, and a binder resin that has a polyolefin resin with a cyclic structure, polynorborene, and a styrene-acrylate resin. Applicants respectfully disagree.

Specifically, Applicants' invention is directed to, *inter alia*, a toner for developing an electrostatically charged copier or printer image using a heat roller fixing means, the toner consisting essentially of a binder resin, a colorant and a charge control agent, wherein the binder resin includes a polyolefin resin having a cyclic structure, wherein the polyolefin resin is a copolymer derived from an alpha-olefin, an alicyclic compound having a double bond and, optionally, a diene monomer, and wherein the electrostatically charged copier or printer image is fixed using a heat fixing means.

It is respectfully submitted that a two-prong inquiry must be satisfied in order to ground a Section 102 rejection. First, the cited reference must contain all of the elements of the claimed invention. *See Lewmar Marine Inc. v. Barient Inc.,* 3 U.S.P.Q.2d 1766 (Fed. Cir. 1987). Second, the reference must contain an enabling disclosure. *See Chester v. Miller,* 15 U.S.P.Q.2d 1333, 1336 (Fed. Cir. 1990). A reference contains an enabling disclosure if a person of ordinary skill in the art could have combined the description of the invention in the reference with his own knowledge of the art to have placed himself in possession of the present invention. *See In re Donohue*, 226, U.S.P.Q. 619, 621 (Fed. Cir. 1985).

Applying the law to the instant facts, the reference relied upon by the Office Action does not disclose, suggest or enable Applicants' invention. First, JP '060 does not contain all of the elements of the instant claims. Claim 16 provides for a compound wherein the polyolefin resin is a copolymer derived from a cyclic olefin copolymer. JP '060 does not teach such a toner. Thus, JP '060 does not read on the instant claims. Second, JP '060 does not enable Applicants' invention wherein the polyolefin resin is a copolymer derived from a cyclic olefin copolymer. Thus, the Section 102 rejection cannot stand and should be obviated.

Consequently, reconsideration and withdrawal of the Section 102 rejections based on JP '060 is respectfully requested.

The Advisory Action also maintained the rejection of the claims under 35 U.S.C. §103(a) as being allegedly unpatentable over Sacripante, U.S. Patent No. 5,324,616 (the "'616 patent") as combined with the Handbook of Imaging Materials, page 169.

The Advisory Action maintained its rejection of the claims under 35 U.S.C. §103(a) as being unpatentable over Japanese Patent 2-184864 ("JP '864") combined with page 13 of Grant & Hackh's Chemical Dictionary.

Finally, the Advisory Action maintained its rejection of the claims under 35 U.S.C. §103(a) as being unpatentable over U.S. 5,817,843 to Masuda ("'843 patent") combined with U.S. 5,741,617 to Inaba ("'617 patent") and U.S. 5,179,171 to Minami ("'171 patent).

These Section 103 rejections will be addressed collectively and are traversed. It is submitted in view of the claim amendments and the remarks herewith, reconsideration and withdrawal of these rejections based on Section 103 are respectfully requested.

The present invention teaches a toner for developing an electrostatically charged image of a heat roller type copier or printer, the toner according to the present invention comprising a binder resin, a colorant and a charge control agent, wherein the binder resin includes a polyolefin resin having a cyclic structure. The polyolefin resin in accordance with the present invention is a copolymer derived from an alpha-olefin, an alicyclic compound having a double bond and, optionally, a diene monomer. The instant invention is also directed to a toner for developing an electrostatically charged copier or printer image using a heat roller fixing means, the toner comprising a binder resin, a colorant and a charge control agent. The binder resin in accordance with the present invention includes a polyolefin resin having a cyclic structure having (i) a low-viscosity resin with a number average molecular weight (Mn) of 1000 to 7500 and a weight average molecular weight (Mw) of 1,000 to 15,000, as measured by GPC, an intrinsic viscosity (i.v.) of less than 0.25 dl/g, and a heat distortion temperature (HDT) by DIN53461-B of lower than 70^0C; and (ii) a high-viscosity resin having a number average molecular weight of at least 7,500 and a weight average molecular weight of at least 15,000, as measured by GPC, an i.v. of 0.25 dl/g or more, and an HDT of 70^0C or higher. The polyolefin resin of the present invention is a copolymer derived from an alpha-olefin, an alicyclic compound having a double bond and, optionally, a diene monomer.

The toner for developing an electrostatically charged image according to the present invention is excellent in fixability, light transmission, and anti-toner spent properties, giving a sharp, high quality image. The toner in accordance with the present invention can apply to any of a dry one-component magnetic toner, a dry one-component nonmagnetic toner, a dry two-component toner and a liquid toner. The toner in accordance with the present invention also exhibits marked effects particularly when used in a color toner.

With respect to JP '864 and Grant & Hackh's Chemical Dictionary, a skilled artisan would readily understand that a polarity control agent is not the same as a charge control agent. A charge control agent ensures stable charge; a polarity control agent does not. Further, the compounds recited in JP '864 would not motivate or lead a skilled artisan to practice the instant invention. To wit, the copolymers discussed in JP '864 are highly complex and different from the alpha-olefins and alicyclic compounds recited in the instant claims. Further, JP '864, at most relates to the combination of two cyclic compounds; not to an alicyclic and an alpha-olefin as instantly claimed. There is no suggestion or motivational teaching that would enable one skilled in the art to develop a toner having a polyolefin resin with a cyclic structure wherein the polyolefin resin is a co-polymer derived from at least an alicyclic compound having a double bond and an alpha-olefin.

Turning to the remaining documents, the '843 patent relates to a quinizarin compounds for dyes; the '617 patent relates to toners having a binder resin, a colorant and a wax composition; and the '171 patent relates to a random copolymer and synthetic waxes. The instant invention, on the other hand, does not use waxes and it is inappropriate to ground the rejection on such disparate references.

It is urged that none of these documents, either alone or in any combination, disclose, suggest, or motivate a skilled artisan to practice the presently claimed invention. In order to ground an obviousness rejection, there must be some teaching which would have provided the necessary incentive or motivation for modifying the reference's teaching. *In re Laskowski*, 12 U.S.P.Q. 2d 1397, 1399 (Fed. Cir. 1989); *In re Obukowitz*, 27 U.S.P.Q. 2d 1063 (B.P.A.I. 1993). Further, "obvious to try" is not the standard under 35 U.S.C. §103. *In re Fine,* 5 U.S.P.Q. 2d 1596, 1599 (Fed. Cir. 1988). And, as stated by the Court in *In re Fritch*, 23 U.S.P.Q. 2d 1780, 1783-1784 (Fed. Cir. 1992): "The mere fact that the prior art may be modified in the manner suggested by the Examiner does not make the modification obvious unless the prior art suggests the desirability of the modification." Also, the Examiner is respectfully reminded that for the Section 103 rejection to be proper, both the suggestion of the claimed invention and the expectation of success must be founded in the prior art, and not Applicants' disclosure. *In re Dow*, 5 U.S.P.Q.2d 1529, 1531 (Fed. Cir. 1988).

In each of the references relied upon in the Advisory Action, the requisite suggestion or motivation is lacking.

Further, and with reference to the '843 patent, Applicants assert that the reference is inappropriate as evidence of obviousness. First, a skilled artisan would not be motivated to practice the instantly claimed cyclic olefin copolymer from the '843 patent's alleged disclosure of an alicyclic hydrocarbon resin. A skilled artisan would consider a cyclic olefin copolymer to be distinguishable from an alicyclic hydrocarbon. Second, even assuming, *arguendo*, the '843 patent discloses that a binder resin can be any binder resin known in the art, that still does not satisfy the legal standard promulgated by the Federal Circuit. It is well-known that a genus disclosed in a reference does not render obvious a species, absent a suggestion or motivation that

would disclose to a skilled artisan the desirability of practicing the species. *See In re Jones*, 958 F.2d 347 (Fed. Cir. 1992). Such a suggestion or motivation is lacking because the '843 patent, *inter alia*, fails to disclose, motivate, teach or suggest the instantly claimed cyclic olefin copolymers.

Further, neither the '617 or '171 patents remedy the inherent deficiencies of the '843 patent. To wit, both the '617 and '171 patents relate to waxes. Specifically, the '617 patent is directed to "toner particles contain[ing] inside the particle a wax composition containing ester wax" (col. 4, lines 32-34). Similarly, the '171 patent is directed to "synthetic waxes" applicable to a laundry list of uses (col. 15, line 53 to col. 16, line 5). The present invention, on the other hand, is not directed to waxes and it is inconceivable that the Advisory Action can come to a different conclusion. Indeed, the '617 and '171 patents teach away from Applicants' invention because a skilled artisan would readily know that when a wax is used as a binder for a toner, adherence of the toner to the substrate (such as paper) is achieved by cold pressure fixing. By contrast, Applicants' invention is directed to a toner for developing an electrostatically charged copier or printer image consisting essentially of a binder resin, a colorant and a charge control agent, wherein the image is fixed by a heat roller fixing means. As no combination of the '843, '617 and '171 patents could lead a skilled artisan to achieve the instantly claimed invention, the rejection is fatally defective.

Accordingly, reconsideration and withdrawal of the Section 103 rejections based on the preceding documents, either alone or in any combination, are respectfully requested.

Any additional fee occasioned by this paper, including with respect to the claim amendments herewith and any fee for a petition for extension of time (to the extent necessary, this paper serves as a petition for an extension of time) to ensure proper pendency of this

application, or any overpayment in those fees, may be charged or credited to Deposit Account No. 50-0320.

In view of the amendments, remarks and attachment herewith, the present application is in condition for allowance. Early and favorable reconsideration and prompt issuance of a Notice of Allowance are earnestly solicited.

If any issue remains as an impediment to allowance, an interview is respectfully urged and the Examiner is further respectfully requested to contact the undersigned by telephone to arrange a mutually convenient time and manner for the interview.

Respectfully submitted,

Frommer Lawrence & Haug LLP
Attorneys for Applicants

By Grace L. Pan
Grace L. Pan
Reg. No. 39,440
Samuel H. Megerditchian
Reg. No. 45,678
TEL: 212 588-0800
FAX: 212 588 0500

VERSION WITH MARKINGS TO SHOW CHANGES MADE

16. (Amended Once) A toner for developing an electrostatically charged copier or printer image [using a heat roller fixing means], the toner consisting essentially of:

a) a binder resin;

b) a colorant; and

c) a charge control agent,

wherein the binder resin includes a polyolefin resin having a cyclic structure, wherein the polyolefin resin is a copolymer derived from an alpha-olefin, an alicyclic compound having a double bond and, optionally, a diene monomer<u>, and wherein the electrostatically charged copier or printer image is fixed using a heat roller fixing means</u>.

17. (Amended Once) A toner for developing an electrostatically charged copier or printer image [using a heat roller fixing means], the toner consisting essentially of:

a) a binder resin;

b) a colorant; and

c) a charge control agent,

the binder resin further comprises a polyolefin resin having a cyclic structure having:

(i) a low-viscosity resin with a number average molecular weight (Mn) of 1000 to 7500 and a weight average molecular weight (Mw) of 1,000 to

15,000, as measured by GPC, an intrinsic viscosity (i.v.) of less than [about] 0.25 dl/g, and a heat distortion temperature (HDT) by DIN53461-B of lower than 70^0C; and

(ii) a high-viscosity resin having a number average molecular weight of at least 7,500 and a weight average molecular weight of at least 15,000, as measured by GPC, an i.v. of [about] 0.25 dl/g or more, and an HDT of 70^0C or higher;

wherein the polyolefin resin is a copolymer derived from an alpha-olefin, an alicyclic compound having a double bond and, optionally, a diene monomer<u>, and wherein the electrostatically charged copier or printer image is fixed using a heat roller fixing means</u>.

18. (Amended Once) A toner for developing an electrostatically charged copier or printer image [using a heat roller fixing means], the toner consisting essentially of:

a) a binder resin;

b) a colorant; and

c) a charge control agent,

the binder resin further comprises a polyolefin resin having a cyclic structure having:

(i) a low-viscosity resin having a number average molecular weight (Mn) of 3,000 to 7,500 and a weight average molecular weight (Mw) of 4,000 to 15,000, as measured by GPC, an intrinsic viscosity (i.v.) of less than

[about] 0.25 dl/g, and a heat distortion temperature (HDT) by DIN53461-B of lower than 70^0C, and

(ii) a high-viscosity resin having a number average molecular weight of 7,500 to 50,000 and a weight average molecular weight of 15,000 to 100,000, as measured by GPC, an i.v. of [about] 0.25 dl/g or more, and an HDT of 70^0C or higher;

wherein the polyolefin resin is a copolymer derived from an alpha-olefin, an alicyclic compound having a double bond and, optionally, a diene monomer, and wherein the electrostatically charged copier or printer image is fixed using a heat roller fixing means.

19. (Amended Once) The toner according to claims 17 or 18, wherein the Mw/Mn ratio, used as a measure of the degree of dispersion of molecular weight distribution, is from [about] 1 to 2.5.

21. (Unchanged) The toner according to claims 16, 17 or 18, wherein the alpha olefin, from which the copolymer is derived, is ethylene.

22. (Amended Once) The toner according to claims 16, 17 or 18, wherein the binder resin includes a polyolefin resin with a cyclic structure having an intrinsic viscosity (i.v.) of [about] 0.25 dl/g or more, a heat distortion temperature (HDT) by DIN53461-B of [about] 70^0C or higher, and a number average molecular weight of 7,500 or more and a weight average molecular weight of 15,000 or more, as measured by GPC, which is contained in a proportion of less than [about] 50% by weight based on the entire binder resin.

23. (Unchanged) The toner according to claims 16, 17 or 18, wherein the binder resin consists of 1 to 100 parts by weight of a polyolefin resin having a cyclic structure, and 0 to 99 parts by weight of at least one resin selected from the group consisting of polyester resins, epoxy resins, polyolefin resins, vinyl acetate resins, vinyl acetate copolymer resins, acrylate resins [and] or styrene-acrylate resins.

24. (Unchanged) The toner according to claims 16, 17 or 18, wherein the polyolefin resin having a cyclic structure [with] comprises at least one functional group selected from the group consisting of a carboxyl group, a hydroxyl group and an amino group.

25. (Unchanged) The toner according to claims 16, 17 or 18, wherein the polyolefin resin having a cyclic structure further comprising a carboxyl group is cross-linked by metal ions or dienes.

Designation: D 648 – 96

Standard Test Method for Deflection Temperature of Plastics Under Flexural Load[1]

This standard is issued under the fixed designation D 648; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

This standard has been approved for use by agencies of the Department of Defense to replace Method 2011 of Federal Test Method Standard 406. Consult the DoD Index of Specifications and Standards for the specific year of issue which has been adopted by the Department of Defense.

1. Scope

1.1 This test method covers the determination of the temperature at which an arbitrary deformation occurs when specimens are subjected to an arbitrary set of testing conditions.

1.2 This test method applies to molded and sheet materials available in thicknesses of 3 mm (1/8 in.) or greater and which are rigid at normal temperature.

NOTE 1—Sheet stock less than 3 mm (0.125 in.) but more than 1 mm (0.040 in.) in thickness may be tested by use of a composite sample having a minimum thickness of 3 mm. The laminae must be of uniform stress distribution. One type of composite specimen has been prepared by cementing the ends of the laminae together and then smoothing the edges with sandpaper. The direction of loading shall be perpendicular to the edges of the individual laminae.

1.3 The values stated in SI units are to be regarded as the standard. The values given in parentheses are for information only.

1.4 *This standard does not purport to address all of the safety concerns, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.*

NOTE 2—This test method, ISO 75-1-1993, and ISO 75-2-1993 are technically equivalent only when the 120 by 10 by 4-mm test specimen is used and deflection temperature is measured at a depth of deflection that is a function of the depth of the specimen, and when the 5-min waiting period to allow specimen creep is applied.

2. Referenced Documents

2.1 *ASTM Standards:*

D 618 Practice for Conditioning Plastics and Electrical Insulating Materials for Testing[2]

D 883 Terminology Relating to Plastics[2]

D 1898 Practice for Sampling of Plastics[2]

E 1 Specification for ASTM Thermometers[3]

2.2 *ISO Standards:*[4]

ISO 75-1-1993 Plastics—Determination of Temperature of Deflection Under Load—Part 1: General Test Method

ISO 75-2-1993 Plastics—Determination of Temperature of Deflection Under Load—Part 2: Plastics and Ebonite

3. Terminology

3.1 *Definitions*—Definitions of plastics used in this test method are in accordance with Terminology D 883 unless otherwise indicated.

4. Summary of Test Method

4.1 A bar of rectangular cross section is tested as a simple beam with the load applied at its center to give maximum fiber stresses of 455 kPa (66 psi) or 1820 kPa (264 psi) (Note 3). The specimen is immersed under load in a heat-transfer medium provided with a means of raising the temperature at 2 ± 0.2°C/min. The temperature of the medium is measured when the test bar has deflected 0.25 mm (0.010 in.). This temperature is recorded as the deflection temperature under flexural load of the test specimen.

NOTE 3—A round robin has been conducted that showed that there is no advantage to using higher loads when measuring deflection temperature of present-day plastics with present-day instruments.[5]

5. Significance and Use

5.1 This test is particularly suited to control and development work. Data obtained by this test method may not be used to predict the behavior of plastic materials at elevated temperatures except in applications in which the factors of time, temperature, method of loading, and fiber stress are similar to those specified in this test method. The data are not intended for use in design or predicting endurance at elevated temperatures.

6. Apparatus

6.1 The apparatus shall be constructed essentially as shown in Fig. 1 and shall consist of the following:

6.1.1 *Specimen Supports*, metal supports for the specimen which shall be 100 mm (4 in.) apart, allowing the load to be applied on top of the specimen vertically and midway between the supports. The contact edges of the supports and of the piece by which load is applied shall be rounded to a radius of 3 mm (1/8 in.). The vertical members which attached the specimen supports to the upper plate shall be

[1] This test method is under the jurisdiction of ASTM Committee D-20 on Plastics and is the direct responsibility of Subcommittee D20.30 on Thermal Properties (Section D20.30.07).

Current edition approved July 10, 1996. Published September 1996. Originally published as D 648 – 41 T. Last previous edition D 648 – 95.

[2] *Annual Book of ASTM Standards*, Vol 08.01.

[3] *Annual Book of ASTM Standards*, Vol 14.03.

[4] Available from American National Standards Institute, 11 W. 42nd St., 13th Floor, New York, NY 10036.

[5] Supporting data giving results of the interlaboratory tests are available from ASTM Headquarters. Request RR: D20-1011.

made of a material having the same coefficient of linear thermal expansion as that used for the rod through which the load is applied.

NOTE 4—Unless these parts have the same coefficient of linear expansion the differential change in length of these parts introduces an error in the reading of the apparent deformation of the specimen. A test should be made on each apparatus using a test bar made of a material having a low coefficient of expansion.[6] The temperature range to be used should be covered and a correction factor determined for each temperature. If this factor is 0.013 mm (0.0005 in.) or greater, its algebraic sign should be noted and the factor should be applied to each test by adding it algebraically to the reading of apparent deflection of the test specimen.

6.1.2 *Immersion Bath*—A suitable liquid heat-transfer medium (Note 5) in which the specimen shall be immersed. It shall be well-stirred during the test and shall be provided with a means of raising the temperature at a uniform rate of 2 ± 0.2°C/min. This heating rate shall be considered to be met if, over every 5-min interval during the test, the temperature of the bath shall rise 10 ± 1°C at each specimen location.

NOTE 5—A liquid heat-transfer medium shall be chosen which will not affect the specimen. Mineral oil is considered safe from ignition to 115°C. Silicone oils may be heated to about 260°C for short periods of time. For still higher temperatures, special heat-transfer media should be used. Improved performance with longer oil life may be obtained by the use of CO_2 or other inert gas to isolate the oil surface from the atmosphere.

NOTE 6—A circulating air oven may be used if it can be shown that equivalent results are obtained.

6.1.3 *Deflection Measurement Device*, suitable for measuring specimen deflection of at least 0.25 mm (0.010 in.). It shall be readable to 0.01 mm (0.0005 in.) or better. The device may be a dial gage or any other indicating or recording device including electric displacement sensing apparatus.

6.1.4 *Weights*—A set of weights of suitable sizes so that the specimen can be loaded to a maximum fiber stress of 1820 kPa (264 psi) ± 2½ % or 455 kPa (66 psi) ± 2½ %. The weight of the rod that applies the testing force shall be determined and included as part of the total load. If a dial gage is used, the force exerted by its spring shall be determined and shall be included as part of the load (Note 7). The load shall be calculated as follows:

$$P = 2Sbd^2/3L$$
$$P' = P/9.80665$$

where:
P = load, N (or lbf),
S = maximum fiber stress in the specimen of 1820 kPa (264 psi) or 455 kPa (66 psi),
b = width of specimen, m (or in.),
d = depth of specimen, m (or in.),
L = width of span between supports (0.1 m (4 in.)), and
P' = load, kgf, when P is in N.

NOTE 7—In some designs of this apparatus the force of the dial gage spring is directed upward and must be subtracted from the load, while in other designs this force acts downward and must be added to the load. Since the force exerted by the spring in certain dial gages varies considerably over the stroke, this force should be measured in that part of the stroke which is to be used. Suggested procedures to determine the total load required to correct for the force of the dial gage spring are given in Appendixes X1 and X2; other procedures may be used if equivalent results are obtained.

6.1.5 *Temperature-Measuring Device*—A thermocouple or thermometer to cover a suitable range. The thermometer shall be one of the following, or its equivalent, as prescribed in Specification E 1: Thermometer 1C, 1F, 2C, or 2F, having ranges from −20 to 150°C, 0 to 302°F, −5 to 300°C, or 20 to 580°F, respectively, whichever temperature range is most suitable for the material being tested.

[6] Invar or borosilicate glass have been found suitable for this purpose.

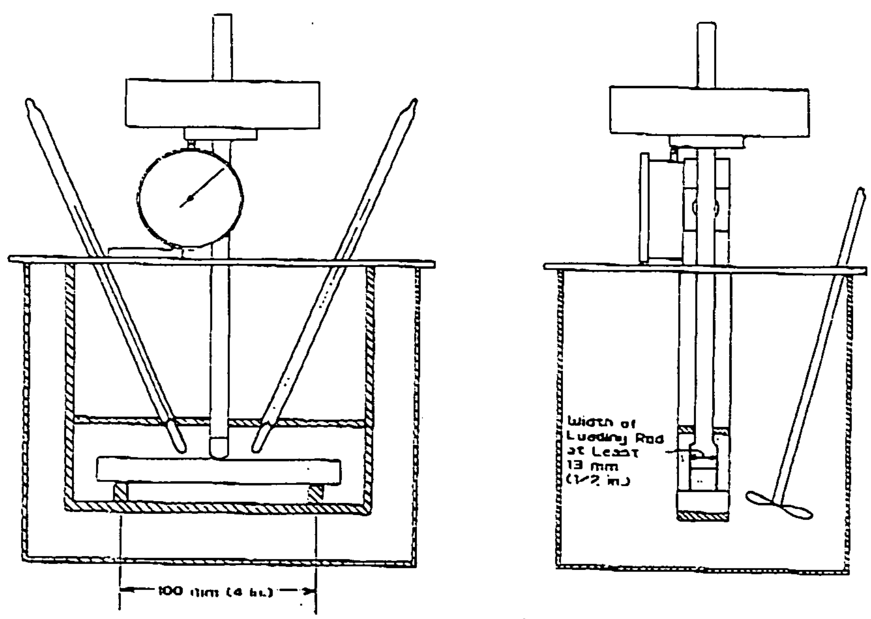

FIG. 1 Apparatus for Deflection Temperature Test

TABLE 1 Statistical Information[A]

Polymer	Average[B] Value	Standard Deviation	Critical[C] Difference, Within Laboratories	Critical[C] Difference, Between Laboratories
Polyethylene, 455 kPa	85.3	4.8	6.0	9.4
Polycarbonate, 455 kPa	142.0	2.0	2.3	3.9
Methyl methacrylate, 1820 kPa	97.6	2.9	4.0	5.7
Polysulfone, 1820 kPa	173.8	2.8	2.3	5.5

[A] All values are given in °C.
[B] Average of pairs.
[C] Between values of a pair.

7. Sampling

7.1 Unless otherwise agreed upon between the seller and the purchaser, sampling shall be in accordance with the General and Specific Sampling Procedures of Practice D 1898. Sampling, based on engineering principles, prior to packaging shall be considered an acceptable alternative.

8. Test Specimens

8.1 At least two test specimens shall be used to test each sample at each fiber stress. The specimen shall be 127 mm (5 in.) in length, 13 mm (½ in.) in depth by any width from 3 mm (⅛ in.) to 13 mm (½ in.). Tolerances on dimensions (for highly reproducible work) should be of the order of ±0.13 mm (0.005 in.) over the length of the specimen.

8.2 The specimens shall have smooth flat surfaces free from saw cuts, excessive sink marks, or flash.

NOTE 8—The type of mold and the molding process used to produce test specimens affect the results obtained in this test method. Cooperating laboratories should therefore standardize the mold and molding procedure to obtain concordant results.

9. Preparation of Apparatus

9.1 The apparatus shall be arranged so that the deflection of the midpoint of the specimen can be measured on a scale calibrated in 0.01 mm (0.005 in.). The apparatus may be arranged to shut off the heat automatically and sound an alarm or record the temperature when the specific deflection has been reached. Sufficient heat transfer liquid shall be used to cover the thermometers to the point specified in their calibration, or 76 mm (3 in.) in the case of the ASTM thermometers referred to in 5.1.5.

10. Conditioning

10.1 *Conditioning*—Condition the test specimens at 23 ± 2°C (73.4 ± 3.6°F) and 50 ± 5 % relative humidity for not less than 40 h prior to test in accordance with Procedure A of Practice D 618, for those tests where conditioning is required. In cases of disagreement, the tolerances shall be ±1°C (±1.8°F) and ±2 % relative humidity.

11. Procedure

11.1 Place the test specimen in the apparatus with its 13-mm (½-in.) dimension vertical. In the case of 13 by 13-mm bars, compression molded or cut from molded sheets, place the bars so that the direction of the testing force will be perpendicular to the direction of the molding pressure.

11.2 Place the temperature measuring device so that it extends to within 3.2 mm (⅛ in.) of the specimen but does not touch it.

11.3 Ascertain that the temperature of the bath is suitable. The bath temperature shall be about room temperature at the start of the test unless previous tests have shown that, for the particular material under test, no error is introduced by starting at a higher temperature.

11.4 Adjust the load so that the desired stress of 1820 kPa (264 psi) ± 2½ % or 455 kPa (66 psi) ± 2½ %, as calculated by the equation of 6.1.4, is obtained.

NOTE 9—Verification of the load should be made on new equipment, after replacement of dial gages, or at any time to ensure that the equipment is in calibration (see Appendixes X1 and X2).

11.5 Adjust the dial gage so that the gage pointer moves upscale at least one-half revolution or 0.64 mm (0.025 in.), whichever is greater when the load is placed on the specimen.

11.6 Five minutes after applying the load, adjust the scale of the gage so that the needle points to zero and start the heating. This waiting period may be omitted when testing materials that show no appreciable creep during the initial 5 min.

NOTE 10—The 5-min waiting period is provided to compensate partially for the creep exhibited by several materials at room temperature when subjected to the prescribed fiber stress. That part of the creep which occurs in the initial 5 min is usually a large fraction of that which occurs in the first 30 min.

11.7 Record the temperature of the medium at which the bar has deflected 0.25 mm (0.010 in.) as the deflection temperature at the specified fiber stress.

NOTE 11—Continuous recording of deflection *versus* temperature, even beyond 0.25 mm (0.010 in.), might be useful in special situations.

12. Report

12.1 Report the following information:

12.1.1 Method of test specimen preparation,

12.1.2 Conditioning procedure,

12.1.3 The width and depth of the specimen, measured to 0.025 mm (0.001 in.),

12.1.4 The deflection temperature and corresponding maximum fiber stress,

12.1.5 The immersion medium and the average heating rate,

12.1.6 Any peculiar characteristics of the specimen noted during the test or after removal from the apparatus, and

12.1.7 Type of tester, automated or manual.

13. Precision and Bias[7]

13.1 *Precision*—An interlaboratory test program was carried out with seven laboratories participating and utilizing both manual and automated instruments. Four polymers were included in the program. Statistical information is summarized in Table 1. The critical difference limits are the limits beyond which observed differences should be considered suspect.

[7] Supporting data are available from ASTM Headquarters. Request RR: D20-1098.

13.2 *Bias*—No statement of bias is possible, since the true value of the property cannot be established by an accepted referee method.

NOTE 12—Analysis of the test data indicates that a bias may exist between manual and automated instruments, with results from manual instruments being an average of 3 to 4°C higher than those from automated instruments.

14. Keywords

14.1 deflection temperature; flexural load; flexure

APPENDIXES

(Nonmandatory Information)

X1. PROCEDURE FOR DETERMINATION OF CORRECT SPECIMEN LOADING UTILIZING EQUILIBRIUM WEIGHING OF THE LOADING ROD

X1.1 Apparatus

X1.1.1 The apparatus shall be constructed essentially as shown in Fig. X1.1 and shall consist of the following:

X1.1.1.1 *Single-Pan or Equal-Arm Laboratory Balance*, having a sensitivity of at least 0.1 g.

X1.1.1.2 *Platform Assembly*, for supporting test unit above the balance.

X1.1.1.3 *Bridge Platform*, for supporting the loading rod on the balance pan.

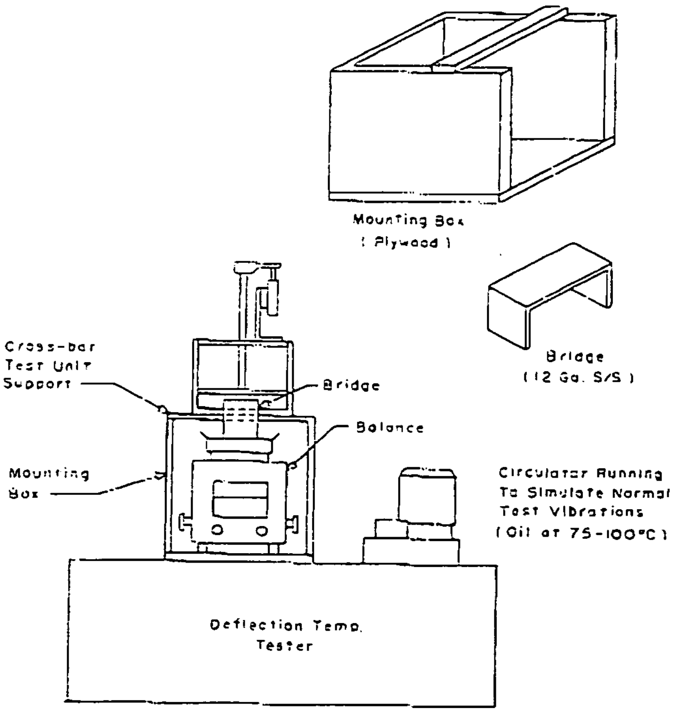

FIG. X1.1 Calibration Apparatus Using Platform Balance

X1.2 Procedure

X1.2.1 Calculate the load required to give the desired fiber stress according to the equation given in 6.1.4.

X1.2.2 Level the mounting assembly on top of the tester (shim or clamp if necessary for firm seating).

X1.2.3 Level the balance.

X1.2.4 Start oil bath stirrer on tester and heat oil to 75 to 100°C and continue operating during calibration.

X1.2.5 Determine tare weight of the bridge.

X1.2.6 Position the test unit on cross bar above balance pan.

X1.2.7 Lubricate the rod and guide hole surfaces with light oil.

X1.2.8 Lift the loading rod and put the bridge in place on the balance pan so that it will support the loading rod (bridge height dimension is such that it supports the rod 13 mm (½ in.) above the level of the specimen supports).

X1.2.9 Adjust the dial face on the dial gage so that the needle points to zero (with no depression of the spindle).

X1.2.10 With the deflector arm in position over the dial gage, lower the rod to the bridge, and then release it very gently. When the balance reaches equilibrium, the desired dial gage movement should be 0.89 ± 0.05 mm (0.035 ± 0.002 in.) (0.64 mm (0.025 in.) as in zero point, plus 0.25 mm (0.010 in.) for deflection of the test bar in the normal test). Read just the deflector arm position until 0.89 ± 0.05 mm is repeatedly obtained at balance.

X1.2.11 Record the force, in grams, at the 0.89 ± 0.05-mm (0.035 ± 0.002-in.) equilibrium deflection.

X1.2.12 Adjust weight of loading rod, or spring force in dial gage, to provide the loading required for a desired stress at 0.89-mm (0.035-in.) deflection according to the equation given in 6.1.4.

NOTE X1.1—The test units (rods, guide surfaces, and dial gage) must be clean and free of any surface imperfections, etc., to achieve precision in calibration and also in normal test use.

X2. PROCEDURE FOR DETERMINATION OF CORRECT SPECIMEN LOADING BY WEIGHING THE APPLIED LOAD WITH A TENSION-TESTING MACHINE

X2.1 Apparatus

X2.1.1 The apparatus shall be constructed essentially as shown in Fig. X2.1 and shall consist of the following:

X2.1.1.1 *Tension-Testing Machine*, of the constant-rate-of jaw separation type, equipped with devices for recording the tensile load and the grip separation. The testing machine used should be capable of measuring loads of at least 2000 g. The rate of separation of the jaws shall be capable of adjustment to 0.51 mm (0.02 in.)/min.

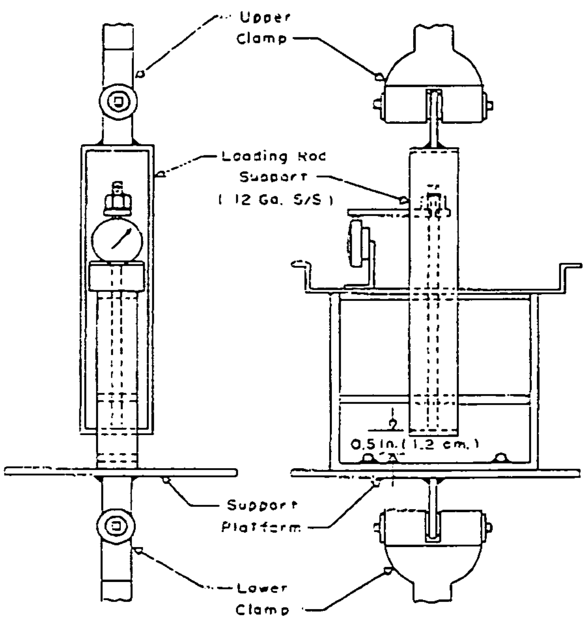

FIG. X2.1 Calibration Apparatus Using a Tensile Machine

X2.1.1.2 *Platform*, square, approximately 203 by 203 mm (8 by 8 in.) to be mounted on the lower crosshead of the tensile machine to support the deflection temperature test unit.

X2.1.1.3 *Loading Rod Support*, a saddle-like device to be clamped in the upper grips of the tensile machine so that it extends under the bottom tip of the loading rod.

X2.2 Procedure

X2.2.1 Mount the support platform in the lower cross-head clamps.

X2.2.2 Fit the loading rod support into the upper clamps and calibrate the tensile-testing machine.

X2.2.3 Secure the deflection temperature test unit on the support platform and adjust the loading rod support so that the tip of the loading rod is 12.7 mm (½ in.) from the top of the specimen supports.

X2.2.4 Lubricate the rod and guide hole surfaces with light oil.

X2.2.5 Adjust the dial gage so that it reads zero, then turn the nut on top of the loading rod clockwise until the deflector arm almost makes contact with the contact arm on top of the dial gage.

X2.2.6 Start the lower crosshead in the up direction at the rate of 0.51 mm (0.02 in.)/min. This in effect causes the loading rod to move down as in an actual test. When the pointer on the dial gage shows movement, activate the chart drive at the rate of 1 in./min.

X2.2.7 Record the force, in grams, at 0.89 ± 0.05-mm (0.035 ± 0.002-in.) deflection.

X2.2.8 Adjust the weight of the loading rod required to give the desired maximum fiber stress according to the equation given in 6.1.4.

X3. PROCEDURE FOR DETERMINATION OF CORRECT SPECIMEN LOADING BY WEIGHING THE APPLIED LOAD IN SITU

X3.1 Scope

X3.1.1 This procedure covers an alternate technique for measuring the net force that is applied to a deflection temperature specimen at midspan.

X3.1.2 The net force is measured with the specimen support unit and loading assembly in place, that is, immersed in the heat-transfer medium.

X3.1.3 This technique permits the user to account for discrepancies in the actual load applied to the specimen as a result of spring forces, friction, buoyancy, etc.

X3.2 Apparatus

X3.2.1 The apparatus shall be constructed essentially as shown in Fig. X3.1 and shall consist of the following:

X3.2.1.1 *Electronic Weighing System with Load Cell* (for example, digital scale or tensile testing machine), single-pan balance, or equal-arm laboratory balance, with a minimum capacity of 2000 g and a sensitivity of 0.1 g.

X3.2.1.2 *Platform Assembly*, for supporting the scale or balance above the deflection temperature bath unit.

X3.2.1.3 *Mass Support Unit*, to hold the loading rod and mass in position while the force measurement is determined.

X3.2.1.4 *Adjustment Fitting*, for connection of the mass support to the load cell or balance. This fitting should facilitate adjusting the test fixture so that the loading force can be measured at the desired position.

X3.3 Procedure

X3.3.1 Determine the loading required to give the desired fiber stress according to the equation given in 6.1.4.

X3.3.2 Place the necessary mass on the loading rod.

X3.3.3 Lower the specimen support unit and loading assembly into the bath.

X3.3.4 Start the circulator, provided that the vibration produced by the circulator motor does not affect the weighing system adversely.

NOTE X3.1—Some vibration from the circulator may be dampened by using rubber feet on the platform assembly, or by designing the

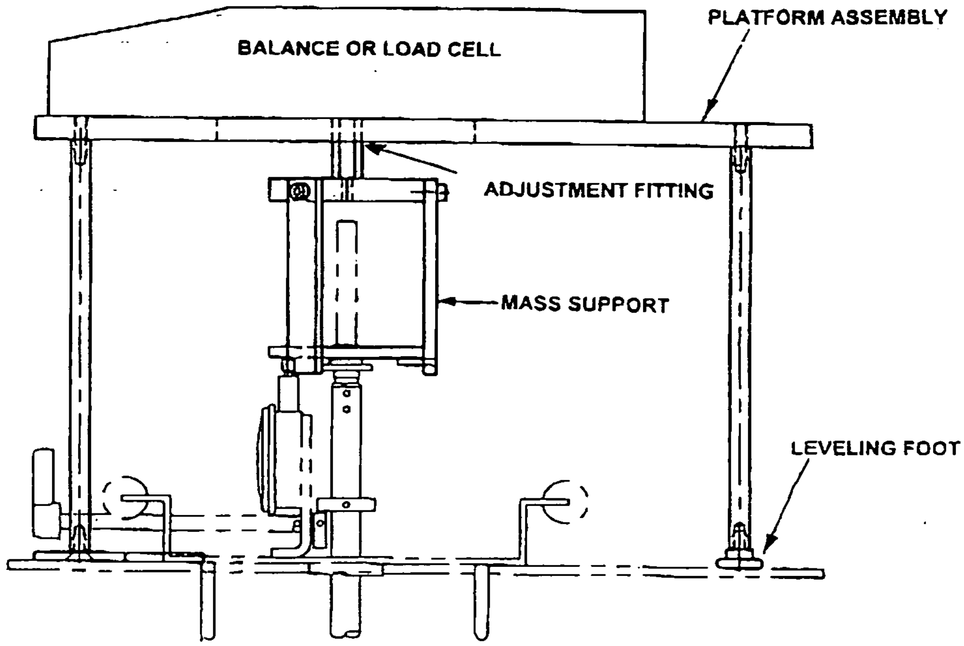

FIG. X3.1 Apparatus for Determination of Correct Specimen Loading

platform assembly so that it spans the bath unit rather than rests on top of it.

X3.3.5 If a scale or balance is used, position the platform assembly on top of the deflection temperature bath unit and level it. Place the scale or balance on top of the platform assembly and verify that it is level.

X3.3.6 Attach the adjustment fitting to the bottom of the load cell or balance.

X3.3.7 Attach the mass support to the bottom of the adjustment fitting.

X3.3.8 If a load cell is used, allow it to warm up before making the measurements. Tare out the weight due to the mass support and adjustment fitting.

X3.3.9 Position the mass support so that it bears the weight of the loading rod and mass.

X3.3.10 Verify that the load cell or balance, adjustment fitting, mass support, and loading rod are uniaxially aligned. It is very important to ensure that the test setup does not introduce any off-center loading into the system that will result in incorrect force measurements.

X3.3.11 Use the adjustment fitting to position the loading assembly so that it corresponds to the zero deflection position. Zero the deflection measurement device of the machine, if necessary. Dial gages should be adjusted in accordance with 11.5.

X3.3.12 Record the indicated load at the zero deflection position to the nearest 0.1 g.

X3.3.13 Use the adjustment fitting to lower the loading assembly to the final deflection position, typically 0.25 mm.

X3.3.14 Record the indicated load at the final deflection point to the nearest 0.1 g.

NOTE X3.2—These force measurements may be made with the bath at any convenient temperature. The effect of temperature on the buoyancy force over the usable range of the machine is generally negligible for commonly used silicone fluids and loading assembly designs. The decrease in the oil density is offset by the increased volume of oil dispersed. If desired, the user may perform this load verification procedure at two different temperatures to confirm the condition.

X3.3.15 Based on these measurements, adjust the mass so that the applied force corresponds to the calculated force of X3.3.1.

X3.3.16 The difference between the force measurement at the zero deflection position (0.00 mm) and the force measurement at the final deflection position (typically 0.25 mm) should be within the ±2.5 % tolerance as specified in 6.1.4.

NOTE X3.3—If the force differential is excessive over the deflection measuring range, the user should attempt to identify the component responsible for the deviation, implement the necessary corrections, and repeat this procedure to ensure that the proper adjustments have been made. It may be possible to adjust the machine so that the calculated load is achieved at an intermediate position (for example, 0.12 mm), thereby permitting the load at the zero deflection position (0.00 mm) and the final deflection position (typically 0.25 mm) to fall within the allowable tolerance.

Designation: D 1601 – 86 (Reapproved 1991)$^{\epsilon 1}$

An American National Standard

Standard Test Method for Dilute Solution Viscosity of Ethylene Polymers[1]

This standard is issued under the fixed designation D 1601; the number immediately following the designation indicates the year of original adoption or, in the case of revision, the year of last revision. A number in parentheses indicates the year of last reapproval. A superscript epsilon (ε) indicates an editorial change since the last revision or reapproval.

ε1 NOTE—Editorial changes were made throughout in March 1991.

1. Scope

1.1 This test method covers the determination of the dilute solution viscosity of ethylene polymers at 135°C. It is applicable to a reasonably wide spectrum of ethylene polymers having densities from 0.910 to 0.970 g/cm^3. Directions are given for the determination of relative viscosity (viscosity ratio), inherent viscosity (logarithmic viscosity number), and intrinsic viscosity (limiting viscosity number).

1.2 The values as stated in SI units are to be regarded as the standard.

1.3 *This standard does not purport to address all of the safety problems, if any, associated with its use. It is the responsibility of the user of this standard to establish appropriate safety and health practices and determine the applicability of regulatory limitations prior to use.*

2. Referenced Documents

2.1 *ASTM Standards:*

D 86 Method for Distillation of Petroleum Products[2]

D 445 Test Method for Kinematic Viscosity of Transparent and Opaque Liquids (and the Calculation of Dynamic Viscosity)[2]

D 1600 Terminology for Abbreviated Terms Relating to Plastics[3]

E 1 Specification for ASTM Thermometers[4]

E 380 Practice for Use of the International System of Units (SI)[5]

3. Terminology

3.1 Units, symbols and abbreviations used in this test method are those recommended in Practice E 380 and Terminology D 1600.

4. Significance and Use

4.1 The knowledge of dilute solution viscosity serves as an additional tool in characterizing ethylene polymers. Viscosity data alone may be of limited value in predicting the processing behavior of the polymer. However, when used in conjunction with other flow and physical property values, the solution viscosity of ethylene polymers may contribute to their characterizations.

4.2 Satisfactory correlation between solution viscosity and certain other properties is possible from polymers of a single manufacturing process. The solution viscosity test is not sensitive to some molecular configurational patterns that may occur among polymers from different manufacturing processes. Hence, its correlation with other properties of polymers produced by different processes, by even one manufacturer, may be limited.

4.3 The viscosity of polymer solutions may be drastically affected by the presence of known or unknown additives in the sample. The use of solution viscosity may be of questionable value where ethylene polymers are known or suspected to contain colorants, carbon black, low molecular weight hydrocarbons, fillers, or other additives.

4.4 The measurement of dilute solution viscosity of ethylene polymers presents problems not ordinarily encountered in viscosimetry. Ethylene polymers are not soluble at room temperature in any known solvent. Some of the higher density materials are insoluble below 100°C. Extreme care must be exercised in transferring the solution to the viscometer for the test if the correct solution concentration is to be maintained. This test has no significance unless the sample is completely soluble.

4.5 The solution viscosity is a function of the root-mean-square size of the polymer molecules in solution. It is known that the solvent selected and the temperature of the determination have an effect on the root-mean-square size of the particles. Hence, where a viscometer, solvent, or temperature other than specified is used, data may not be comparable to that obtained by this procedure.

5. Apparatus

5.1 *Volumetric Flasks*, 100-mL, grade EXAX or better.[6]

5.2 *Transfer Pipets*, grade EXAX or better.[6]

5.3 *Constant Temperature Bath*, capable of maintaining 135 ± 0.1°C.

5.4 *Viscometer*—Modified Ubbelohde viscometer[7] as shown in Figs X1.1, X1.2, and X1.3 and described in Appendix X1.

---

[1] This test method is under the jurisdiction of ASTM Committee D-20 on Plastics and is the direct responsibility of Subcommittee D20.70 on Analytical Methods (Section D20.70.05).

Current edition approved Oct. 31, 1986. Published December 1986. Originally published as D 1601 – 58. Last previous edition D 1601 – 78$^{\epsilon 1}$.

[2] *Annual Book of ASTM Standards*, Vol 05.01.

[3] *Annual Book of ASTM Standards*, Vol 08.01.

[4] *Annual Book of ASTM Standards*, Vol 14.03.

[5] *Annual Book of ASTM Standards*, Vol 14.02.

[6] Glassware used in this test method should be tested in accordance with the procedures described in the National Institute of Standards and Technology Circular No. C-434, "Testing of Glass Volumetric Apparatus," and should not exceed the limits of accuracy set forth in the circular.

[7] The modified Ubbelohde viscometer is available from J. V. Stabin, 84-21 Midland Parkway, Jamaica, Queens, New York, NY, 11431, and Cannon Instrument Co., P.O. Box 812, State College, PA, 16801.

NOTE 1—Other types of viscometers may be used provided they can be shown to agree with the type specified.

5.5 *Oven*, maintained at 140 ± 5°C.

5.6 *Timer*, as specified in 4.5 of Test Method D 445.

5.7 *Thermometer*—An ASTM High Softening Point Thermometer having a range from 30 to 200°C, and conforming to the requirements for Thermometer 16C in Specification E 1.

6. Reagents and Materials

6.1 *Solvent—Decahydronaphthalene*, practical grade, purified and redistilled, as follows:

6.1.1 The solvent shall be purified by percolation through 100 to 200 mesh commercial grade silica gel. This treatment removes naphthalene, tetrahydronaphthalene, and oxy compounds, particularly peroxides.

6.1.2 The redistilled product shall conform to the following requirements when tested in accordance with Method D 86:

Standard Distillation	ASTM Method D 86
Initial boiling point	190°C min
10 mL	191°C min
20 mL	192°C min
80 mL	194°C max
90 mL	195°C max
Dry point	196°C max

NOTE 2—While use of other solvents, such as tetrahydronaphthalene or xylene, may sometimes be advantageous, they will generally yield different values for solution viscosities.

6.1.3 Immediately after redistillation of the decahydronaphthalene, 0.05 to 0.1 % by weight of an antioxidant may be added to inhibit oxidation during the viscosity determination. Phenyl betanaphthylamine and 4-methyl-2,6-ditertiary-butylphenol have been found satisfactory. There is evidence to show that stabilization of the solvent with one of these antioxidants renders its solutions with most ethylene polymers resistant to oxidation at 135°C for periods of many hours.

6.2 *Heat Transfer Medium*—Any liquid heat transfer medium that will not appreciably affect the accuracy of the test may be used. Care should be exercised in using fluids that discolor or smoke with prolonged heating.

NOTE 3—The following materials have been used: (*1*) Silicone fluid,[8] that has been found to be free of discoloration at 135°C for sustained periods. Some silicone fluids are not miscible with others, (*2*) Peanut oil, (*3*) Primol D, a mineral oil. Peanut oil and Primol D oil may darken on prolonged heating; however, this darkening may be inhibited through the use of antioxidants.

7. Sampling

7.1 Ordinarily, ethylene polymers are readily soluble to decalin at 135°C. Where solubility is desired in minimum time the sample shall be pulverized to pass a 20-mesh screen.

NOTE 4—The sample can be conveniently pulverized in a rotary cutting mill with a 20-mesh screen fitted to its pulverizing chamber. The Wiley mill, containing blades on its rotor which impact the material against stationary blades fitted into the pulverizing chamber walls, has been found satisfactory. It is sold by a number of scientific supply houses. Care must be taken to prevent overheating unusually hard or tough polymer samples during pulverizing. Such overheating can possibly cause thermal degradation.

8. Conditioning

8.1 The sample need not be predried or conditioned, unless previous specific knowledge of the sample makes it advisable. Ordinarily, ethylene polymers containing no additives are not hygroscopic.

9. Measurement of Solvent Viscosity

9.1 Determine the efflux time of the solvent in the viscometer in accordance with 10.6 through 10.9 until three consecutive efflux times agree within 0.2 s. The average of these readings shall be used in calculating the relative viscosity of the solution, as described in Section 11.

NOTE 5—The solvent viscosity may be calculated from the efflux time in accordance with the following equation:

$$V = Ct - B/t$$

where:

V = kinematic viscosity, cSt,
C = calibration constant for the viscometer,
t = efflux time, s, and
B = kinetic energy and end effect correction constant. (This constant is dependent on the design of the viscometer and may be determined experimentally, along with the constant C, by use of suitable calibrating fluids. In general, with viscometers designed so that B is relatively small, the correction B/t is negligible for relatively long efflux times.)

NOTE 6—Although the kinetic energy effect inherent in the design of the viscometer may be determined by Test Method D 445, that method is believed to be tedious for liquids with viscosities in the range of decalin at 135°C. The following equations provide a more convenient and precise means of calculating the relative viscosity (viscosity ratio), η_r, corrected for the kinetic energy contribution.

$$\eta_r = \frac{t_s + \Delta t_s}{t_o + \Delta t_o} = \frac{t_s - D/t_s}{t_s - D/t_o}$$

where:

t_s and t_o = flow times of the solution and solvent, respectively,
Δt = kinetic energy correction term for a particular liquid, and
D = a constant for the viscometer.

D and Δt may be calculated as follows:

$$D/t = (-Vm\rho)/L\eta 8\pi = \Delta t$$

where:

t = flow time of the liquid in the viscometer,
V = efflux volume of the liquid,
m = kinetic energy correction coefficient. (If the viscometer capillary has trumpet-shaped ends that break away rather sharply, and Δt is no larger than 3 % of t, it is probably safe to assume m equal to unity.)
ρ = density of the liquid,
L = length of the capillary, and
η = viscosity of the liquid.

Using this equation, the viscometer constant D, may be calculated after the following measurements are made:

(*1*) Flow time of a liquid (water for example) of known

[8] The silicone fluids available from the Dow Corning Corp., Midland, MI, or from the Union Carbide Corp., Linde Silicones Div., New York, NY, have been found satisfactory for this purpose.

viscosity and density in the viscometer,

(2) Capillary length, and

(3) Volume of the viscometer bulb. This may be done by filling the bulb with water and measuring the volume of the water in a graduated cylinder. An accuracy of ±3 % is adequate for all measurements, if Δt is no larger than 3 % of t. This will provide a precision of better than 0.1 %.

10. Procedure

10.1 If it is desired to determine only the relative viscosity (viscosity ratio) or the inherent viscosity (logarithmic viscosity number), weigh one specimen of 0.18 to 0.22 ± 0.0002 g and transfer it quantitatively to a 100-mL volumetric flask which has been purged with nitrogen. ~ 0,2 %

NOTE 7—Usually the determination of relative or inherent viscosity (viscosity ratio or logarithmic viscosity number) may suffice.

10.2 To determine the intrinsic viscosity (limiting viscosity number), weigh four specimens of the following approximate weights to ± 0.0002 g:

Specimen 1	0.09 to 0.11 g
Specimen 2	0.18 to 0.22 g
Specimen 3	0.28 to 0.32 g
Specimen 4	0.38 to 0.42 g

NOTE 8—The intrinsic viscosity (limiting viscosity number) may also be measured using successive dilutions of only one, or duplicate, solutions. This has the advantage of requiring that only one polymer specimen be weighed. Dilutions to the solution of this specimen in the viscometer are a time-saving device. The technique is as follows: Weigh one, or a duplicate, specimen of 0.38 to 0.42 g to an accuracy of ±0.0002 g and transfer it quantitatively to a 100-mL volumetric flask, which has been purged with nitrogen, using a funnel and washing the watch glass and funnel down with solvent. Place the specimen into solution in accordance with 10.3 through 10.5 and measure the relative viscosity (viscosity ratio) in accordance with 10.6 through 10.9.

Separate dilutions of the original solution to desired concentrations can be made by adding solvent at 135°C in precisely measured quantities to the original solution in the viscometer and the relative viscosity (viscosity ratio) at each dilution tested before the next dilution is made. For example, the following technique might be used.

First Dilution—Add *exactly* 5 mL of solvent at 135°C to the filter stick of the viscometer and calculate the solution concentration from the known weight of solute and total volume of solvent in the viscometer. (This concentration should be approximately 0.3 g/100 mL.) Mix the solution in the viscometer thoroughly by pulling it into the viscometer bulb and back again three times. Determine the relative viscosity (viscosity ratio) at this concentration.

Second Dilution—Add *exactly* 10 mL of solvent at 135°C and calculate the concentration, as above. (This concentration should be approximately 0.2 g/100 mL.) Determine the relative viscosity (viscosity ratio) at this concentration.

Volumetric limitations of the modified Ubbelohde viscometer require that approximately 15 mL of solution be added to the instrument for the first determination of relative viscosity (viscosity ratio). Also, the viscometer will not hold and properly mix liquid volumes very much in excess of 30 mL. Hence, use of the above procedure will provide only three values of relative viscosity (viscosity ratio) from which intrinsic viscosity (limiting viscosity number) can be determined. Other dilution techniques may be used to obtain four or more values of relative viscosity (viscosity ratio), but the range of solute concentrations is limited unless the reverse procedure is used—that of introducing a minimum volume of solvent into the viscometer to obtain solvent viscosity, then adding aliquots of a concentrated stock solution to the viscometer to obtain successive values of relative viscosity (viscosity ratio) at higher concentrations.

10.3 Add approximately 50 mL of solvent to the specimen flasks, purge them again with nitrogen, stopper them loosely and place them in an oven or bath maintained at 140°C. Vent the flasks at the end of 10 min. Shake the flasks once every 10 min until the solution is complete (Notes 9 and 11).

NOTE 9—Most ethylene polymers will go into complete solution at the solute concentrations used in this method within 2 h. Some polymer samples may require up to 4 h. If the solute has not dissolved in 6 h, this procedure may not be applicable, due to possibly incomplete solution of the sample not detectable to the eye. If incomplete solution of the sample is suspected, the viscometer filter stick may be removed from the instrument after the sample solution has been transferred and the filter stick dried under vacuum. Any weight difference after drying should show evidence of undissolved residue. Even with the use of an antioxidant in the solvent, significant oxidation of the polymer sample may occur in unusually long dissolving period.

NOTE 10—**Caution**—When the solution is considered complete, examine visually to be certain that no undissolved particles, gels, or particles of foreign matter are present.

10.4 Place the solutions in the volumetric flasks in the constant temperature bath maintained at 135 ± 0.1°C. Dilute the solutions to approximately 99 mL with the solvent maintained at the bath temperature, and thoroughly mix the solutions by hand. Replace the flasks in the bath.

10.5 After ½ h, complete the dilution to the 100-mL mark with the solvent maintained at 135°C by means of a transfer pipet and shake the flasks again.

NOTE 11—Be certain the solutions are uniformly mixed. Do not allow excessive cooling while shaking the flasks. The error due to expansion of the flask at 135°C is believed not to be significant in this test.

10.6 Cut about 0.5 in. from the lower tip of a transfer pipet to permit faster transfer of its contents to the viscometer. Place the pipet in a 140°C oven for 30 min. It is preferable that the pipet be fitted with a suitable heating mantel to retard precipitation of the polymer on its walls during transfer of the solution (Note 12). Purge the viscometer thoroughly with nitrogen gas using a slow, gentle stream. Fill the pipet from the sample solution flask and transfer approximately 15 mL of the solution into the filter stick of the modified Ubbelohde viscometer which is permanently positioned in the bath (Note 13). (See Appendix X1 for directions for use and cleaning of the viscometer.)

NOTE 12—A graduate cylinder previously heated in the viscosity bath may be used instead of a pipet. The hot solution can be poured into the graduate positioned in the bath, the graduate removed after about 3 min, and its contents poured into the viscometer filter stick.

NOTE 13—If the dilution method described in Note 8 is used, *exactly* 15 mL of solution must be transferred to the viscometer, using the transfer pipet described in 10.6.

10.7 Bring the liquid level to approximately 10 mm above the upper graduation mark on the viscometer capillary.

10.8 Allow the solution to drain to the upper mark on the capillary. As the meniscus passes this point, start the timer and time the interval for the solution to drain to the lower mark on the capillary.

10.9 Measure the efflux time of the solution at least three times. Three consecutive readings shall agree within 0.2 s.

11. Calculation

11.1 *Relative Viscosity* (*Viscosity Ratio*)—Calculate the relative viscosity (viscosity ratio) for each concentration measured, from the average efflux times as follows:

$$\eta_r = t/t_o$$

where:

η_r = relative viscosity (viscosity ratio)
t = average efflux time of solution, and
t_o = average efflux time of pure solvent.

11.2 *Inherent Viscosity* (*Logarithmic Viscosity Number*)—Calculate the inherent viscosity (logarithmic viscosity number) for each concentration measured, as follows:

$$\eta_{inh} = \ln \eta_r/C$$

where:

η_{inh} = inherent viscosity (logarithmic viscosity number) at concentration C, and
$\ln \eta_r$ = natural logarithm of the relative viscosity (viscosity ratio), and
C = concentration in grams/100 mL of solution.

11.3 *Intrinsic Viscosity* (*Limiting Viscosity Number*)[9]—Plot the four logarithmic viscosity numbers *versus* their respective concentrations on rectilinear graph paper. Draw the best straight line through the points and extrapolate it to zero concentration. The intrinsic viscosity (limiting viscosity number), [η], is the intercept of the line at zero concentration.

NOTE 14—If desired, the specific viscosity (η_{sp}) and the reduced viscosity (viscosity number) (η_{red}) may be calculated as follows:

$$\eta_{sp} = \eta_r - 1$$
$$\eta_{red} = \eta_{sp}/C$$

NOTE 15—As a check on the plot of the intrinsic viscosity (limiting viscosity numbers) *versus* their concentrations, the specific viscosity (η_sp) divided by their concentrations for the four specimens may be plotted *versus* their concentrations on the same graph. The slopes of these two lines will not be the same, but they should converge to the same value at zero concentration. This additional line serves to more accurately position the zero concentration point.

12. Report

12.1 The report shall include the following:

12.1.1 Complete identification of the material tested including type, source, manufacturer's code numbers, and trade name.

12.1.2 Conditioning procedure used, if any.

12.1.3 The relative viscosity (viscosity ratio) of one or more concentrations, depending on whether it is desired to obtain relative, inherent, or intrinsic viscosity (viscosity ratio, logarithmic viscosity number, or limiting viscosity number).

12.1.4 The intrinsic viscosity (limiting viscosity number), when desired, to three significant figures.

13. Precision and Bias

13.1 A task group is currently being formed in D20.70.05 to provide precision and bias data.

14. Keywords

14.1 ethylene polymers; dilute solution viscosity

[9] Schulken, R. M., and Sparks, M. L., *Journal of Polymer Science*, JPSCA, Vol XXVI, 1957, p. 227.

APPENDIX

(Nonmandatory Information)

X1. DIRECTIONS FOR USE AND CLEANING OF THE MODIFIED UBBELOHDE VISCOMETER

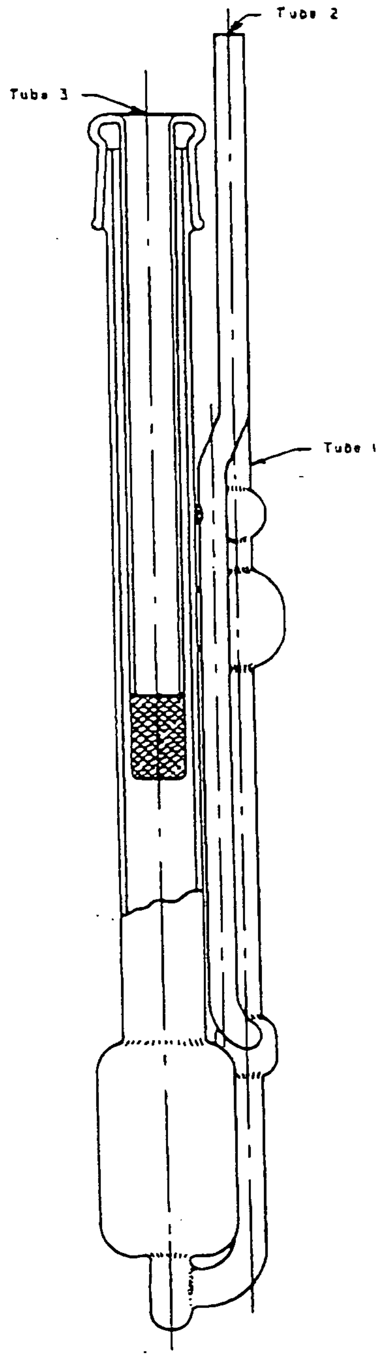

FIG. X1.1 Modified Ubbelohd Viscometer

X1.1 Using Viscometer

X1.1.1 Pour the hot solution immediately into the viscometer through tube 3 containing the filter stick, as shown in Figs. X1.1 and X1.2. Slip a rubber tube with one end closed off by a stopcock over tube 2 of the viscometer. Cautiously apply air or nitrogen pressure, using a pressure controller, in filling tube 3 in order to force the solution through the glass filter. Remove the pressure controller. Remove excess solution from the pores of the filter by applying the end of the intake tube of a suction flask to the filter surface; this is necessary in order to prevent the formation of an air trap in the system. Care must be taken in the use of suction in cleaning the filter to avoid loss of sample.

X1.1.2 Allow the solution to stand in the bath for at least 5 min to ensure temperature equilibration. Keep the stopcock on tube 2 closed, and cautiously apply pressure to tube 3, forcing the liquid into viscometer tube 1 until it fills the small upper bulb. Then release the pressure and open the stopcock on tube 2 to the atmosphere allowing the solution to flow freely out of the capillary tube. Measure the time, in seconds, required for the meniscus to pass from the upper to the lower mark on tube 1. Repeat the determinations until three successive efflux times agree within 0.2 s; the average of these three readings is used for calculating the viscosity.

X1.2 Cleaning Viscometer

X1.2.1 Empty the viscometer after each determination without removing it from the 135°C bath. Remove the filter stick (Fig. X1.3) from the viscometer and insert the intake tube of the suction flask into tube 3 so that its lower end touches the bottom of the viscometer tube. Apply a vacuum to the suction flask and withdraw the solution. Rinse the viscometer in place with fresh hot solvent. Measure the efflux time of pure solvent after every determination. If the value obtained differs by more than ± 0.3 s from the value previously obtained in the clean viscometer, further cleaning is necessary.

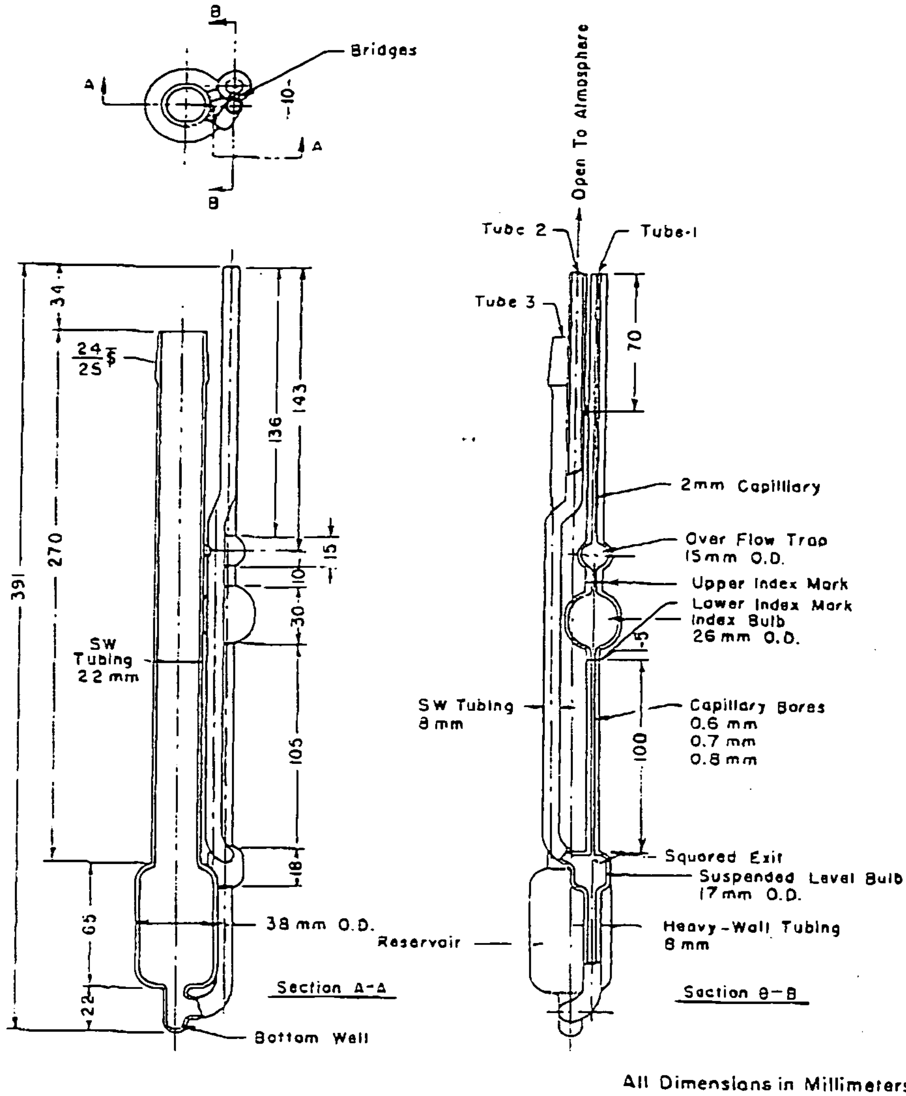

FIG. X1.2 Section Views of the Modified Ubbelohde Viscometer

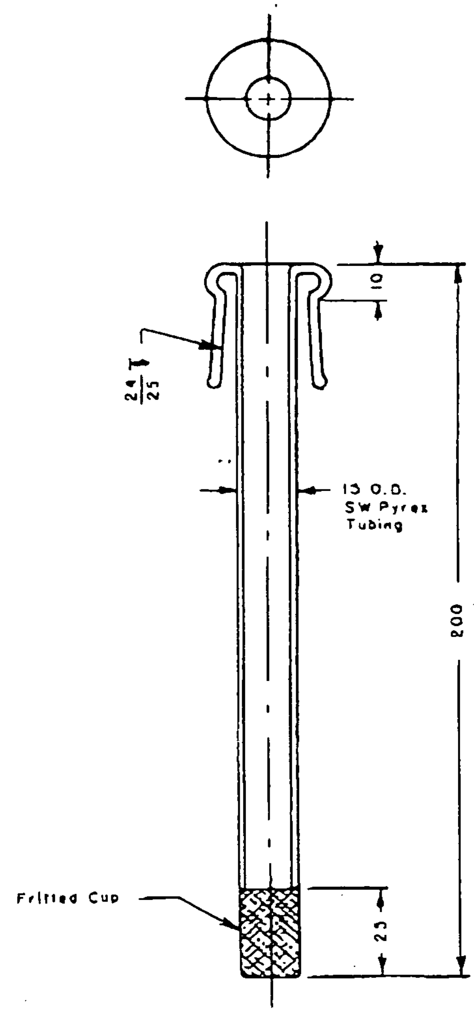

FIG. X1.3 Filter Stick of the Modified Ubbelohde Viscometer

The American Society for Testing and Materials takes no position respecting the validity of any patent rights asserted in connection with any item mentioned in this standard. Users of this standard are expressly advised that determination of the validity of any such patent rights, and the risk of infringement of such rights, are entirely their own responsibility.

This standard is subject to revision at any time by the responsible technical committee and must be reviewed every five years and if not revised, either reapproved or withdrawn. Your comments are invited either for revision of this standard or for additional standards and should be addressed to ASTM Headquarters. Your comments will receive careful consideration at a meeting of the responsible technical committee, which you may attend. If you feel that your comments have not received a fair hearing you should make your views known to the ASTM Committee on Standards, 1916 Race St., Philadelphia, PA 19103.

INTERNATIONAL STANDARD

ISO 75-2

First edition
1993-09-15

Plastics — Determination of temperature of deflection under load —

Part 2:
Plastics and ebonite

Plastiques — Détermination de la température de fléchissement sous charge —

Partie 2: Plastiques et ébonite

Reference number
ISO 75-2:1993(E)

Plastics — Determination of temperature of deflection under load —

Part 2:
Plastics and ebonite

1 Scope

1.1 This part of ISO 75 specifies three methods for the determination of the temperature of deflection under load (bending stress) of plastics and ebonite:

— method A, using a nominal surface stress of 1,80 MPa;

— method B, using a nominal surface stress of 0,45 MPa;

— method C, using a nominal surface stress of 8,00 MPa.

1.2 The test specimens are tested in one of two positions, flatwise or edgewise, the requirements on test-specimen dimensions being different in each case (see clause 6).

1.3 See ISO 75-1:1993, subclause 1.3.

NOTE 1 The methods give better reproducibility with amorphous plastics than with semi-crystalline ones. With some materials, it may be necessary to anneal the test specimens to obtain reliable results. Annealing procedures, if applied, generally result in an increase in the temperature of deflection under load (see 6.2 and 6.3).

2 Normative references

The following standards contain provisions which, through reference in this text, constitute provisions of this part of ISO 75. At the time of publication, the editions indicated were valid. All standards are subject to revision, and parties to agreements based on this part of ISO 75 are encouraged to investigate the possibility of applying the most recent editions of the standards indicated below. Members of IEC and ISO maintain registers of currently valid International Standards.

ISO 75-1:1993, *Plastics — Determination of temperature of deflection under load — Part 1: General test method.*

ISO 293:1986, *Plastics — Compression moulding test specimens of thermoplastic materials.*

ISO 294:—[1], *Plastics — Injection moulding of test specimens of thermoplastic materials.*

ISO 2818:—[2], *Plastics — Preparation of test specimens by machining.*

ISO 3167:1993, *Plastics — Multipurpose test specimens.*

3 Definitions

See ISO 75-1:1993, clause 3.

4 Principle

A standard test specimen made of plastic or ebonite is subjected to a bending stress to produce one of the nominal surface stresses given in 1.1. The temperature is raised at a uniform rate, and the temperature at which a specified deflection occurs is measured.

1) To be published. (Revision of ISO 294:1975)

2) To be published. (Revision of ISO 2818:1980)

8.2 Initial temperature of the heating equipment

See ISO 75-1:1993, subclause 8.2.

8.3 Measurement

See ISO 75-1:1993, subclause 8.3.

Apply one of the nominal surface stresses specified in 8.1 of this part of ISO 75.

Note the temperature at which the bar reaches the standard deflection given in table 1 or 2 for the test-specimen height concerned (thickness h for specimens tested in the flatwise position and width b for specimens tested in the edgewise position). This temperature is the temperature of deflection under load.

NOTES

4 The initial flexural strain due to the loading of the specimen at room temperature is neither specified nor measured in these methods. The specified quantity, the standard deflection s, is essentially a deflection difference, corresponding to a flexural-strain difference. The ratio of this flexural-strain difference to the initial flexural strain depends on the modulus of elasticity, at room temperature, of the material under test. This method is not suitable, therefore, for comparing the temperatures of deflection under load of materials with widely differing elastic properties.

5 The standard deflections given in tables 1 and 2 correspond to a flexural strain of 0,2 % at the surface of the test specimen.

9 Expression of results

See ISO 75-1:1993, clause 9.

If the individual results for amorphous plastics or ebonite differ by more than 2 °C, or those for semi-crystalline materials by more than 5 °C, repeat tests shall be carried out.

Table 1 — Standard deflection for different test-specimen heights — 80 mm × 10 mm × 4 mm specimen tested in the flatwise postion

Test-specimen height (thickness h of specimen) mm	Standard deflection mm
3,8	0,36
3,9	0,35
4,0	0,34
4,1	0,33
4,2	0,32

Table 2 — Standard deflection for different test-specimen heights — 120 mm × (3,0 to 4,2) mm × (9,8 to 15,0) mm specimen tested in the edgewise position

Test-specimen height (width b of specimen) mm	Standard deflection mm
9,8 to 9,9	0,33
10,0 to 10,3	0,32
10,4 to 10,6	0,31
10,7 to 10,9	0,30
11,0 to 11,4	0,29
11,5 to 11,9	0,28
12,0 to 12,3	0,27
12,4 to 12,7	0,26
12,8 to 13,2	0,25
13,3 to 13,7	0,24
13,8 to 14,1	0,23
14,2 to 14,6	0,22
14,7 to 15,0	0,21

10 Precision

See ISO 75-1:1993, clause 10.

11 Test report

See ISO 75-1:1993, clause 11.

The information on

h) the orientation of the test specimen (flatwise or edgewise);

i) the nominal surface stress;

may be given as follows:

Use method A, B or C to designate the nominal surface stress and the letters "e" and "f" to designate the test-specimen orientation.

Thus a test using a nominal surface stress of 1,80 MPa and flatwise test-specimen orientation would be referred to as "method Af". Similarly, a test using a nominal surface stress of 0,45 MPa and edgewise orientation would be called "method Be".

INTERNATIONAL STANDARD ISO 1628-3 : 1991
TECHNICAL CORRIGENDUM 1

Published 1992-02-01

Plastics — Determination of viscosity number and limiting viscosity number —

Part 3: Polyethylenes and polypropylenes

TECHNICAL CORRIGENDUM 1

Plastiques — Détermination de l'indice de viscosité et de l'indice limite de viscosité —
Partie 3: Polyéthylènes et polypropylènes
RECTIFICATIF TECHNIQUE 1

Technical corrigendum 1 to International Standard ISO 1628-3 : 1991 was prepared by Technical Committee ISO/TC 61, *Plastics*, Sub-Committee SC 9, *Thermoplastic materials.*

Page 4

Subclause 8.1

Replace the second equation by the following:

$$c = \frac{m}{V \cdot \gamma}$$

and the line following this equation (on page 5) by

"m, V and γ having the same meaning as in 7.4.1."

UDC 678.742.2/.3 : 532.13 Ref. No. ISO 1628-3 : 1991/Cor.1 : 1992 (E)

Descriptors: plastics, polymers, polyethylene, polypropylene, tests, determination, viscosity.

INTERNATIONAL STANDARD

ISO 1628-3

First edition
1991-04-01

Plastics — Determination of viscosity number and limiting viscosity number —

Part 3:
Polyethylenes and polypropylenes

Plastiques — Détermination de l'indice de viscosité et de l'indice limite de viscosité —

Partie 3: Polyéthylènes et polypropylènes

Reference number
ISO 1628-3:1991(E)

Plastics — Determination of viscosity number and limiting viscosity number —

Part 3:
Polyethylenes and polypropylenes

1 Scope

This part of ISO 1628 defines particular conditions for determining the viscosity number and limiting viscosity number of polyethylenes and polypropylenes at 135 °C in dilute solution. It is applicable to low, medium and high-density polyethylenes and to a wide range of polypropylenes, including pure isotactic, stereoblock and atactic polymers.

The viscosity of polymer solutions may be affected by additives present in the sample. The value of a viscosity number determined by this method may therefore be unreliable if the sample contains fillers or other additives.

NOTE 1 Viscosity number is also known as the Staudinger function (J_v) and limiting viscosity number as the Staudinger index (J_g).

2 Normative references

The following standards contain provisions which, through reference in this text, constitute provisions of this part of ISO 1628. At the time of publication, the editions indicated were valid. All standards are subject to revision, and parties to agreements based on this part of ISO 1628 are encouraged to investigate the possibility of applying the most recent editions of the standards indicated below. Members of IEC and ISO maintain registers of currently valid International Standards.

ISO 1628-1:1984, *Guidelines for the standardization of methods for the determination of viscosity number and limiting viscosity number of polymers in dilute solution — Part 1: General conditions.*

ISO 3105:1976, *Glass capillary kinematic viscometers — Specification and operating instructions.*

3 Principle

The times of flow of a solvent and a solution of polymer at a specified concentration in that solvent are measured at 135 °C. The viscosity number and limiting viscosity number are calculated from these measurements and from the known concentration of the solution.

Ethylene and isotactic polypropylene polymers are not soluble at room temperature in any known solvents. Precautions must therefore be taken during the test to avoid any precipitation of polymer, which would give an incorrect solution concentration.

4 Solvent

4.1 **Decahydronaphthalene**, analytical reagent grade, redistilled at a temperature not higher than 65 °C and a pressure of approximately 500 Pa; its peroxidation is prevented by suitable means, for example distilling in the presence of hydroquinone.

Immediately after redistillation, 0,2 % (*m*/*m*) of antioxidant shall be added to inhibit oxidation during the viscosity determination. Antioxidants which have been found satisfactory include:

- 4,4'-thio-bis-(6-*tert*-butyl-3-methyl)phenol;
- bis-(2-hydroxy-3-*tert*-butyl-5-methyl)phenylmethane;
- octadecyl-3-(3',5'di-*tert*-butyl-4'-hydroxyphenyl) *n*-propionate;
- and tetrakis[methylene 3-(3',5'di-*tert*-butyl-4'-hydroxyphenyl) *n*-propionate]methane.

Dimensions in millimetres

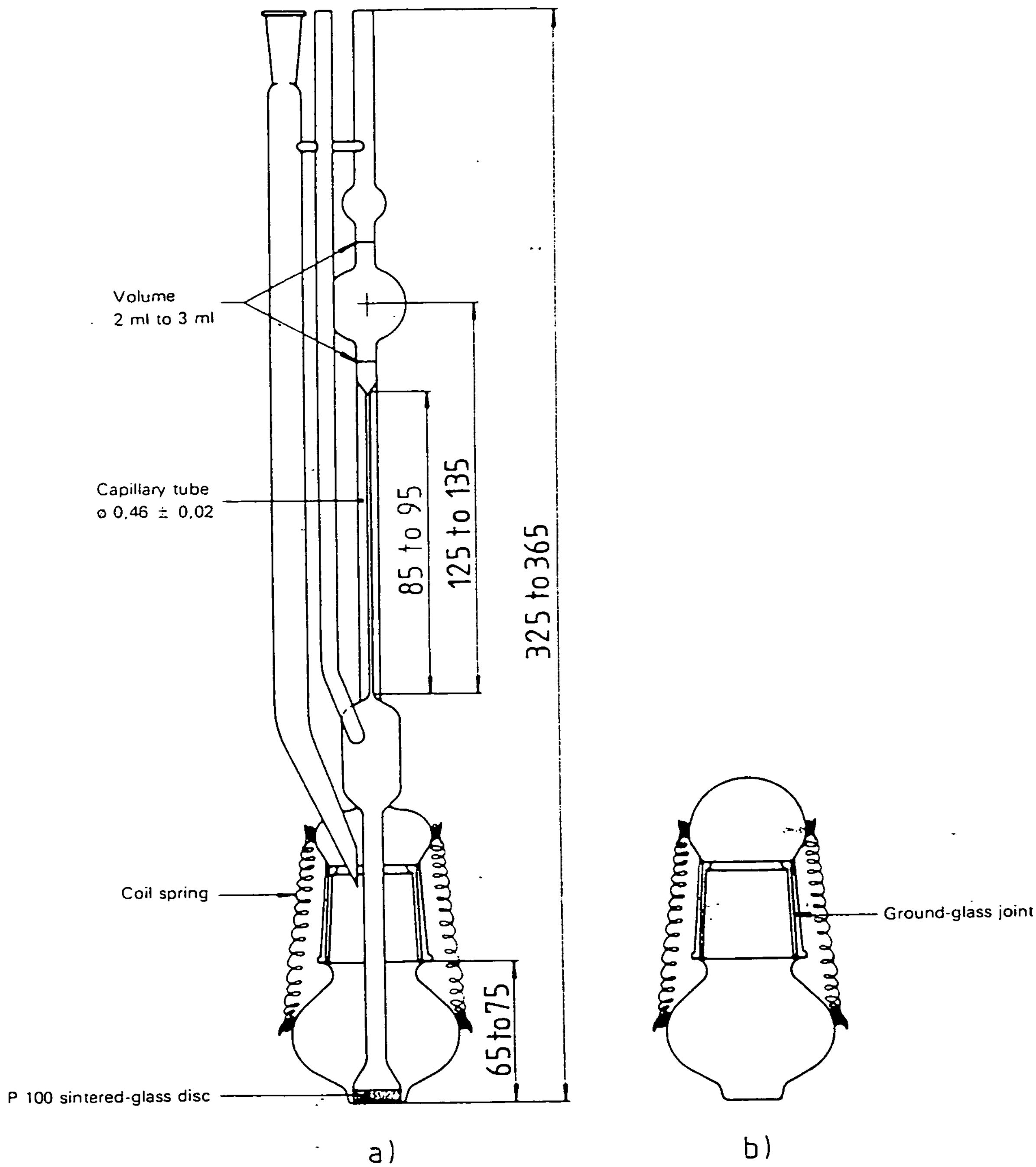

Figure 1 — Ubbelohde viscometer modified for high-temperature tests

m, V, γ and ρ_{20} having the same meaning as in 7.4.1.

The calculated value shall be rounded off to the nearest whole number.

8.2 Limiting viscosity number, $[\eta]$

Calculate the limiting viscosity number $[\eta]$, expressed in millilitres per gram, using the equation

$$[\eta] = \frac{VN}{1 + k \cdot c \cdot VN}$$

where

k is a coefficient depending on polymer concentration and polymer structure;

c and VN have the same meaning as in 8.1.

An approximate value of $[\eta]$ can be calculated using $k = 0{,}27$ but, in the case of precise determinations, k has to be determined for each range of concentrations and for each series of samples.

The calculated value shall be rounded off to the nearest whole number.

8.3 Precision

The precision of this test method is not known because inter-laboratory data are not available. This method may not be suitable for use in specifications or in case of any disputed results as long as these data are not available.

9 Test report

The test report shall include the following particulars:

a) a reference to this part of ISO 1628;

b) all details necessary for the complete identification of the material tested (type, origin, manufacturer's trade mark);

c) any treatment given to the sample before testing (cutting, grinding, etc.);

d) if applicable, the part of a manufactured article tested (whole article, surface or interior);

e) the dissolution time, if this exceeds 2 h;

f) the viscosity number, giving in parentheses the concentration used, for example: VN(0,001) = 380 ml/g;

g) the limiting viscosity number, giving in parentheses the constant k calculated for the same concentration.